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Extracting buried twists with polarized neutron reflectometry

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Abstract. Polarized neutron reflectometry can extract the depth-dependent magnetization of thin-film magnets more precisely than other techniques, which are limited to a measurement of the average moment or the moment at the surface. Measuring the reflectivity first with neutrons glancing off the front surface and again with neutrons glancing off the back surface yields eight spin cross sections instead of the usual four. Differences in the front and back reflectivities indicate the presence of magnetic twists in the sample. We have applied this method, as well as the more conventional front-surface-only reflectometry, to study a soft ferromagnet exchange-coupled to a hard ferromagnet in fields from 5 to 50 mT.

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Polarized neutron reflectometry (PNR) gives the researcher a powerful technique by which to study the magnetization of thin films. Other methods, such as bulk magnetometry, magneto-optical indicator film (MOIF) imaging, magneto-optic Kerr effect (MOKE) and X-ray magnetic circular dichroism (XMCD), have some drawbacks. The first two techniques are limited to measuring only the average magnetization of the sample; they cannot extract the magnetic structure of constituent layers of thin films. All but the first technique are constrained by the penetration depth of photons in magnetic material: sensitivity to magnetic layers buried tens of nanometers deep inside the structure is reduced. Cold-neutron reflectometry uses neutrons with wavelengths comparable to the length-scale over which the magnetism varies in artificial magnetic structures. The scattering process gives a depth-sensitivity not attainable with imaging techniques. PNR (see, e.g., [1] for a general quantum mechanical description) extracts the component of the magnetization M parallel or perpendicular to the polarization vector $\boldsymbol{p}$. Using this information, the projection of the vector magnetization M onto the surface can be found as a function of depth, with a vertical resolution $l \sim 0.1$ nm.

The growth of the magnetic recording industry has spurred the creation of a number of artificial magnetic structures [2], including exchange-spring magnets [3]. The magnetic behavior of many of these structures is not yet fully understood; hence researchers have turned to PNR to study how the spins in these materials reorient under conditions of varying magnetic field and temperature.

In the typical PNR experiment, neutrons are polarized along a quantization axis $+z$, typically by a supermirror [4]. A flipper placed before the sample can invert the polarization of this incident beam when necessary. Another flipper and polarizing element are placed in the exit beam to select the orientation of the detected neutron. Thus, four reflectivities $R^{ij} = R^{++}$, R^{--}, R^{+-}, or R^{-+} can be measured. The index i indicates the polarization of the incident beam, while j indicates the polarization of the detected beam. The first two reflectivities are the non-spin-flip (NSF) while the last two are known as spin-flip (SF). The nuclear potential always contributes to the NSF reflectivity.

Inset (a) of Fig. 1 shows the geometry of a reflectivity experiment. The polarization $\boldsymbol{p} \parallel z$ lies perpendicular to the scattering vector $\boldsymbol{q} \parallel y$. The NSF reflectivity is strongly sensitive to the component of magnetization $M_{||} \parallel z$, while the SF reflectivity is strongly sensitive to $M_{\perp} \parallel x$. In this geometry, $R^{+-} = R^{-+}$, and the SF reflectivities cannot distinguish whether $\boldsymbol{p} \times M_{\perp}$ is parallel or antiparallel to q. Hence for non-collinear magnetic structures, a measurement of R^{+-} does not identify whether the moments tend to twist clockwise or counterclockwise. Consequently, it is often difficult to establish with certainty whether the magnetic structure is non-collinear, or whether it is collinear with the direction of magnetization at an arbitrary angle with respect to the polarization axis. By orienting $\boldsymbol{p} \parallel q$, all magnetic scattering appears in the SF channels, and (for, e.g., single-domain helical samples), a difference between R^{+-} and R^{-+} identifies them as non-collinear.

In our recent work [5] with $\boldsymbol{p} \perp q$, we reported that with two measurements of reflectivity one can distinguish whether some structures are non-collinear. In the first measurement

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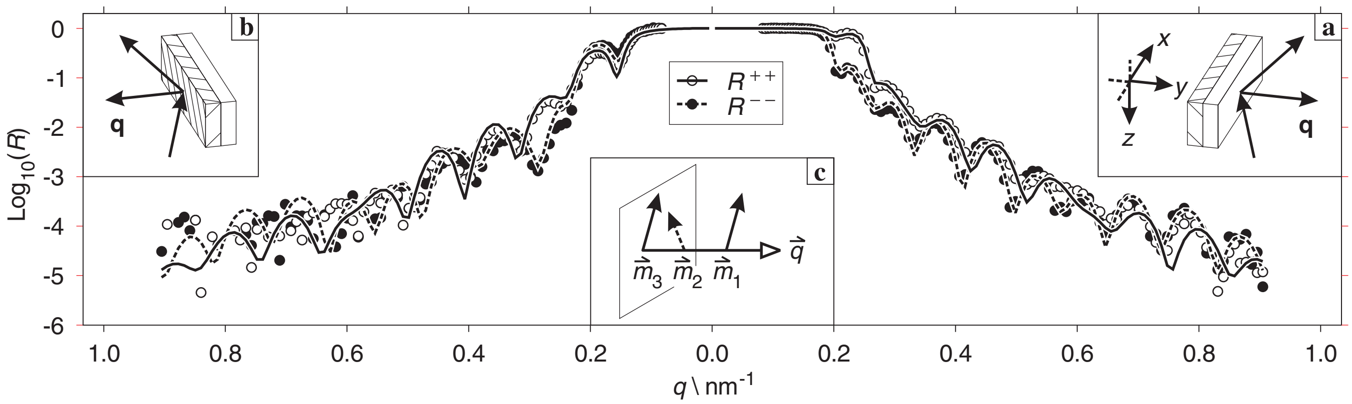

Fig. 1a–c. Non-spin-flip reflectivity of an exchange-spring magnet under 26 mT. The data are plotted with *circles*, and the fits with the *lines*. The errorbars are negligble when $\boldsymbol{q} < 0.62\,\mathrm{nm}^{-1}$. **a** Geometry of scattering from the front of a bilayer. Neutrons are polarized along z. The surface normal is parallel to y and the scattering vector $\boldsymbol{q}$. **b** Geometry of scattering from the back. Only $\boldsymbol{q}$ has changed. **c** Orientation of layer moments and $\boldsymbol{q}$ in a trilayer film with a mirror plane at m_2

neutrons scatter off the front face of the sample ($q \parallel y$, as in Fig. 1a). In the second the neutrons scatter off the back of the sample ($\boldsymbol{q} \parallel -y$, as in Fig. 1b). In some cases, mere inspection of the eight measured cross-sections is sufficient to identify non-collinear structures. A simultaneous fitting of them quickly eliminates alternate structures which might otherwise be plausible given only the first measurement. The main portion of Fig. 1 shows the front/back NSF reflectivity of an exchange-coupled film exhibiting exchange-spring behavior. The bilayer film consists of $Fe_{0.55}Pt_{0.45}$(20 nm) topped by $Ni_{0.80}Fe_{0.20}$(50 nm) with capping and seed layers of Pt and a substrate of glass. The front reflectivity is plotted on the right, with $\boldsymbol{q}$ increasing towards the right. The back reflectivity is plotted on the left with $\boldsymbol{q}$ increasing towards the left. Prior to the measurement a magnetic field of 890 mT was applied along $-z$. The data were collected in a field of 26 mT along $+z$. The fits to the data reveal a structure with a smooth twist of 90°, of which 48° lies in the hard ferromagnet FePt. Details can be found in [5]. There is a clear difference in the front/back reflectivities near the critical edge (below which the NSF reflectivity is identically 1). We shall now discuss the key features responsible for the observed asymmetry of actual magnetic films by exploring the reflectivity of simple models.

1 Model calculations

To calculate the reflectivity, we use the exact solution to the Schrödinger equation, with a formalism nearly identical to that of Rühm [6]. To help us extract the essential features of the magnetic structure responsible for the back/front asymmetry, we first make some rather dramatic simplifications. To eliminate effects of refraction, which must occur when neutrons scatter off the back of a magnetic film deposited on a substrate, we consider only free-standing films—ones without a substrate. We set the nuclear scattering length density $bN = 0$ to extract only the magnetic portion of the cross-section. We model a two-layer magnetic film in which the only difference between the layers is the direction of the magnetization m_j. The magnitude of magnetization and thickness of the layers are identical, but the angle from the net magnetization M to m_1 is θ, and from M to m_2 is $-\theta$. The angle from z to M is Θ. The polarization $\boldsymbol{p}$ is parallel to z and the scattering vector $\boldsymbol{q} \parallel y$ is normal to the surface of the film. In this paper we focus on the front/back asymmetry of the NSF scattering. Analogous examples and arguments can be made for the SF reflectivity. The vector relationships are shown in Fig. 2a and the angular relationships are shown in Fig. 2b.

Figure 2a shows the NSF reflectivity for our simplified structure when $\Theta = 0$ and $\theta \neq 0$. The "front reflectivity" is that measured when neutrons encounter m_1 first and "back reflectivity" is that measured when neutrons encounter m_2 first. On each side is a cartoon describing the orientation of $\boldsymbol{q}$, $\boldsymbol{p}$ and the moments m_j. In this geometry, the polarization $\boldsymbol{p}$ bisects the two layer magnetizations and there is no net moment

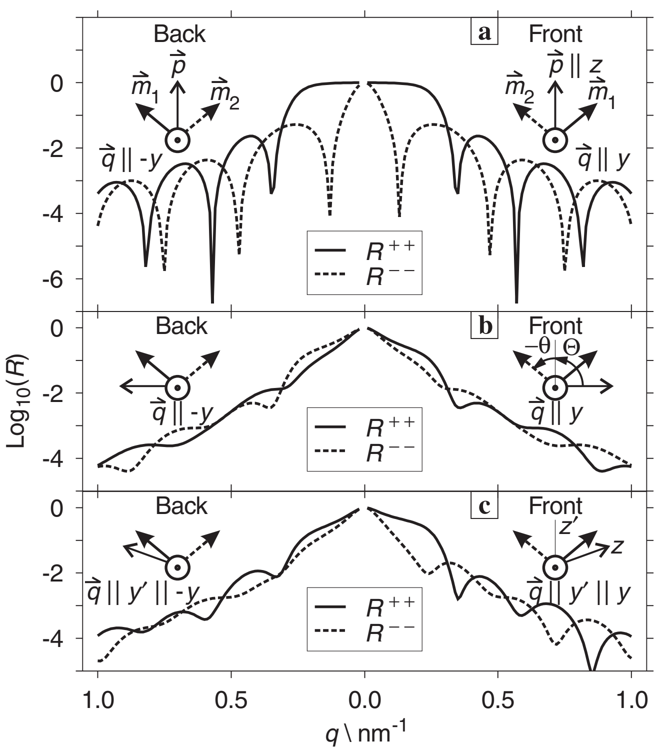

Fig. 2. a Non-spin-flip reflectivity from a model bilayer with polarization $\boldsymbol{p}$ bisecting the angle between the layer moments m_1 and m_2. Neutrons strike m_1 first on the right and m_2 first on the left. **b** Non-spin-flip reflectivity of the structure in a) with the polarization axis $\boldsymbol{p}$ rotated 90° from that picture. All *arrows* reference the same labels as before. **c** Non-spin-flip reflectivity of the structure in (a) with the polarization $\boldsymbol{p}$ at an "arbitrary" angle. The natural coordinate system y' and z' are explained in the text

perpendicular to $\boldsymbol{p}$. Because our scattering geometry is insensitive to the sign of the x component of the magnetization, we expect the front and back reflectivities to be identical, as is evident from the figure. The splitting between R^{++} and R^{--} indicates that there is a net moment in the film.

We rotate the sample (or alternatively, $\boldsymbol{p}$) about $\boldsymbol{q}$ so that $\Theta = 90°$ and show the resulting reflectivity in Fig. 2b. With zero net moment projected along $\boldsymbol{p}$, the two NSF reflectivities interchange when switching from front to back reflectivity. The splitting between R^{++} and R^{--} results from $\theta \neq 0$. When we collapse the non-collinear structure to a collinear one with $\theta = 0$, the splitting vanishes and the front and back reflectivities are identical, and, not surprisingly, are very nearly the average of the NSF reflectivities in Fig. 2b.

In Fig. 2c we give an intermediate value to Θ. As expected, the front and back reflectivities are intermediate between identical and interchanged. Near the critical $\boldsymbol{q}$ there is a pronounced asymmetry between front and back which immediately identifies this structure as non-collinear. Real samples have nonvanishing nuclear scattering length density bN and include a substrate. If we add only the nuclear potential to our model, the reflectivity plotted in Fig. 2c changes to closely resemble that of our actual data in Fig. 1. The nuclear and substrate contributions induce a small additional asymmetry on top of the dominant magnetic contribution.

2 Discussion

The front/back asymmetry can be understood in the invariant vector derivation of the reflectivity of Rühm et al. [6]. The computation of the reflectivity of a multilayer medium is straightforward, but involves the multiplication of numerous (4×4) transfer matrices A_j which describe the propagation of the neutron wave function through a homogenous magnetic layer j. The difference in computing front and back reflectivity is the order in which the A_j are multiplied. The sample transfer matrix $A = \prod_j A_j$ yields [6] the (2×2) reflectance matrix $\hat{R} = \frac{1}{2}(R_0 I + \boldsymbol{R} \cdot \boldsymbol{\sigma})$ where I is the identity matrix and $\boldsymbol{\sigma}$ is the vector of the Pauli spin matrices. $\boldsymbol{R}$ is a three-component vector of complex numbers and is nonzero for magnetic media. The NSF reflectivity is given by the expression $R^{\mathrm{NSF}} = \frac{1}{4}|R_0 + \boldsymbol{R} \cdot \boldsymbol{p}|^2$ where $\boldsymbol{p}$ is the polarization of the neutrons.

The simplified bilayer film we have been considering in Fig. 2 imposes a natural Cartesian coordinate system $\{x', y', z'\}$ in which the net magnetization M lies along z' and the surface normal y' is coincident with $\boldsymbol{q}$ of the lab frame in Fig. 1. (Fig. 2c shows the relation among y, y', z and z'.) For small θ, we can compute Taylor's expansion of $\boldsymbol{R}$ about $\theta = 0$ which gives $\boldsymbol{R} = \theta R_x \hat{\boldsymbol{x}}' + \theta R_y \hat{\boldsymbol{y}}' + R_z \hat{\boldsymbol{z}}'$ in which R_x, R_y and R_z have no dependence on θ. To understand the front/back asymmetry we need to understand how $\hat{R}$ transforms when we interchange layers 1 and 2. An inspection of the magnetization diagram in Fig. 2b shows that the interchange $1 \to 2$ is equivalent to the transformation $\theta \to -\theta$, so that

$$R^{\mathrm{NSF}}_{\mathrm{front}} = \tfrac{1}{4}|R_0 + \theta R_x p_{x'} + \theta R_y p_{y'} + R_z p_{z'}|^2 \quad \text{and}$$

$$R^{\mathrm{NSF}}_{\mathrm{back}} = \tfrac{1}{4}|R_0 - \theta R_x p_{x'} - \theta R_y p_{y'} + R_z p_{z'}|^2 \,. \tag{1}$$

These equations indicate an alternative interpretation. We see that the back reflectivity with polarization $\boldsymbol{p}$ is identical to the front reflectivity obtained when $\boldsymbol{p}$ is rotated 180° about z'. Thus when $\boldsymbol{p} \parallel \hat{\boldsymbol{z}}'$ ($\Theta = 0$), the reflectivity is unchanged and when $\boldsymbol{p} \perp \hat{\boldsymbol{z}}'$ ($\Theta = 90°$) the reflectivities interchange. For other values of Θ the back/front reflectivity provides two different (but symmetry related) polarization axes from the one axis defined in the laboratory frame.

The symmetry of our model is quite low. At the interface between layers 1 and 2 there is a two-fold axis of rotation along the mean magnetization M. We can replace this with a mirror plane by placing a third layer, identical to layer 1, on the other side of layer 2, as sketched in Fig. 1c. Now the transformation (front) $\to$ (back) is equivalent to the interchange of layers 1 and 3. But the interchanged structure is identical to the original structure, so $R^{\mathrm{NSF}}_{\mathrm{front}} = R^{\mathrm{NSF}}_{\mathrm{back}}$.

We have seen how non-collinear magnetic structures with no mirror planes parallel to the front surface can exhibit an asymmetry between back and front reflectivities when the net magnetization M makes a non-zero angle Θ with respect to the polarization axis $\boldsymbol{p}$. The Born approximation is inadequate to explain this effect.

References

1. G.P. Felcher, R.O. Hilleke, R.K. Crawford, J. Haumann, R. Kleb, G. Ostrowski: Rev. Sci. Instrum. **58**, 609 (1987)
2. P. Grünberg: Physics Today **54**, 31 (2001)
3. E.F. Kneller, R. Hawig: IEEE Trans. Magn. **27**, 3588 (1991)
4. C.F. Majkrzak: Physica B **221**, 342 (1996)
5. K.V. O'Donovan, J.A. Borchers, C.F. Majkrzak, O. Hellwig, E.E. Fullerton: Phys. Rev. Lett. **88**, 067 201 (2002)
6. A. Rühm, B.P. Toperverg, H. Dosch: Phys. Rev. B **60**, 16 073 (1999)